AFRL-RY-WP-TP-2013-0180

LONG-WAVELENGTH INFRARED SURFACE PLASMONS ON Ga-DOPED ZnO FILMS EXCITED VIA 2D HOLE ARRAYS FOR EXTRAORDINARY OPTICAL TRANSMISSION (PREPRINT)

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OCTOBER 2013
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UNITED STATES AIR FORCE

REPORT DOCUMENTATION PAGE

Form Approved
OMB No. 0704-0188

1. REPORT DATE ***(DD-MM-YY)***
October 2013

2. REPORT TYPE
Journal Article Preprint

3. DATES COVERED ***(From - To)***
1 October 2011 – 1 July 2013

4. TITLE AND SUBTITLE
LONG-WAVELENGTH INFRARED SURFACE PLASMONS ON Ga-DOPED ZnO FILMS EXCITED VIA 2D HOLE ARRAYS FOR EXTRAORDINARY OPTICAL TRANSMISSION (PREPRINT)

5a. CONTRACT NUMBER
In-house

5b. GRANT NUMBER

5c. PROGRAM ELEMENT NUMBER
61102F

6. AUTHOR(S)
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5d. PROJECT NUMBER
3001

5e. TASK NUMBER
12

5f. WORK UNIT NUMBER
Y00S

7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES)
Optoelectronic Technology Branch
Aerospace Components & Subsystems Division
Air Force Research Laboratory, Sensors Directorate
Wright-Patterson Air Force Base, OH 45433-7320
Air Force Materiel Command, United States Air Force

Solid State Scientific Corporation
Wyle Laboratories

8. PERFORMING ORGANIZATION REPORT NUMBER
AFRL-RY-WP-TP-2013-0180

9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES)
Air Force Research Laboratory , Sensors Directorate
Wright-Patterson Air Force Base, OH 45433-7320
Air Force Materiel Command
United States Air Force

10. SPONSORING/MONITORING AGENCY ACRONYM(S)
AFRL/RYDH

11. SPONSORING/MONITORING AGENCY REPORT NUMBER(S)
AFRL-RY-WP-TP-2013-0180

12. DISTRIBUTION/AVAILABILITY STATEMENT
Approved for public release; distribution unlimited.

13. SUPPLEMENTARY NOTES
Journal article to be published in SPIE Proceedings, Volume 8809, Plasmonic Propagation. ©2013 SPIE. The U.S. Government is joint author of the work and has the right to use, modify, reproduce, release, perform, display or disclose the work.PAO Case Number 88ABW-2013-3490, Clearance Date 7 August 2013. Document contains color.

14. ABSTRACT
Extraordinary optical transmission (EOT) through highly conductive ZnO films with sub-wavelength hole arrays is investigated in the long-wavelength infrared regime. EOT is facilitated by the excitation of surface plasmon polaritons (SPPs) and can be tuned utilizing the physical structure size such as period. Pulse laser deposited Ga-doped ZnO has been shown to have fluctuations in optical and electrical parameters based on fabrication techniques, providing a complimentary tuning means. The sub-wavelength 2D hole arrays are fabricated in the Ga-doped ZnO films via standard lithography and etching processes. Optical reflection measurements completed with a microscope coupled FTIR system contain absorption resonances that are in agreement with analytical theories for excitation of SPPs on 2D structures. EOT through Ga-doped ZnO is numerically demonstrated at wavelengths where SPPs are excited. This highly conductive ZnO EOT structure may prove useful in novel integrated components such as tunable biosensors or surface plasmon coupling mechanisms.

15. SUBJECT TERMS
plasmonics, infrared, EOT, doped zinc oxides

16. SECURITY CLASSIFICATION OF:			17. LIMITATION OF ABSTRACT:	18. NUMBER OF PAGES	19a. NAME OF RESPONSIBLE PERSON (Monitor)
a. REPORT	b. ABSTRACT	c. THIS PAGE			Justin W. Cleary
Unclassified	Unclassified	Unclassified	SAR	10	19b. TELEPHONE NUMBER (Include Area Code) N/A

Long-wavelength infrared surface plasmons on Ga-doped ZnO films excited via 2D hole arrays for extraordinary optical transmission

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ABSTRACT

Extraordinary optical transmission (EOT) through highly conductive ZnO films with sub-wavelength hole arrays is investigated in the long-wavelength infrared regime. EOT is facilitated by the excitation of surface plasmon polaritons (SPPs) and can be tuned utilizing the physical structure size such as period. Pulse laser deposited Ga-doped ZnO has been shown to have fluctuations in optical and electrical parameters based on fabrication techniques, providing a complimentary tuning means. The sub-wavelength 2D hole arrays are fabricated in the Ga-doped ZnO films via standard lithography and etching processes. Optical reflection measurements completed with a microscope coupled FTIR system contain absorption resonances that are in agreement with analytical theories for excitation of SPPs on 2D structures. EOT through Ga-doped ZnO is numerically demonstrated at wavelengths where SPPs are excited. This highly conductive ZnO EOT structure may prove useful in novel integrated components such as tunable biosensors or surface plasmon coupling mechanisms.

Keywords: plasmonics, infrared, EOT, doped zinc oxides.

1. INTRODUCTION

Surface plasmon polaritons (SPPs) are a means of real-time, label-free biosensing [**1-3]** with current commercial surface plasmon resonance (SPR) biosensors being based in the visible or near-infrared wavelength regimes. Mid- to long-wave infrared (LWIR) SPR based biosensors would take advantage of the characteristic vibrational modes of biomolecules in this wavelength regime potentially giving increased sensitivity. SPPs also have potential in the development of mid- to long-wave infrared plasmonic waveguides or even hybrid dielectric/plasmonic subsystems [**4**] that could lead towards “plasmonic circuit” integration.

Extraordinary optical transmission (EOT) is a phenomenon in which transmission through a 2D periodic structure is larger than thought possible by classical aperture theory. This phenomenon is largely correlated with the excitation of SPPs on 2D periodic structures although this is not the complete explanation [**5**]. EOT has been a recent field of interest with such 2D structures expanding the possible on-chip applications of surface plasmons [**5-6**]. LWIR SPPs have been characterized on 2D structures fabricated in doped-Si, InAs, and GaAs [**7-9**], as well as Cu coated Ni mesh [**10**]

with the doped-GaAs and Cu/Ni systems being specifically investigated for EOT. The doped semiconductors 2D structures may have promise for on-chip plasmonics and/or EOT applications with doping tunability playing a vital role.

The investigation of alternative plasmonic materials has recently been accelerated [**4, 8, 11-22**] due to the large losses expected when using noble metals as plasmon hosts and the drive to better enable fabrication of on-chip systems using standard techniques. Aluminum and gallium doped zinc oxides have been proposed as possible low-loss plasmonic materials in the near-IR [**12, 14-15**] and mid- to long-wave IR [**22**]. ZnO work has demonstrated largely fluctuating free carrier concentration, as well as mobility, [**14, 23-25**] with slight changes in fabrication techniques and stoichiometry. These material properties ultimately are responsible in determining SPP properties such as propagation length and penetration depth [**22**], which are important in the aforementioned applications and have been investigated recently for hybrid plasmonic-photonic waveguides [**26-27**]. Doped ZnO then has potential and should be considered for a novel class of broadly tunable plasmonic host materials.

Investigated here are 2D-hole array coupled plasmons in the mid- and long-wave infrared for EOT purposes. Presented are theoretical and experimental investigations pertaining to ZnGaO as surface plasmon host materials. Fabricated structures, characterized via reflection measurements and finite element simulations, are utilized to demonstrate potential EOT properties. Periodic hole array structures for EOT are shown in Figure 1 with pattern periods being P_x and P_y illustrating the x- and y-directions respectively.

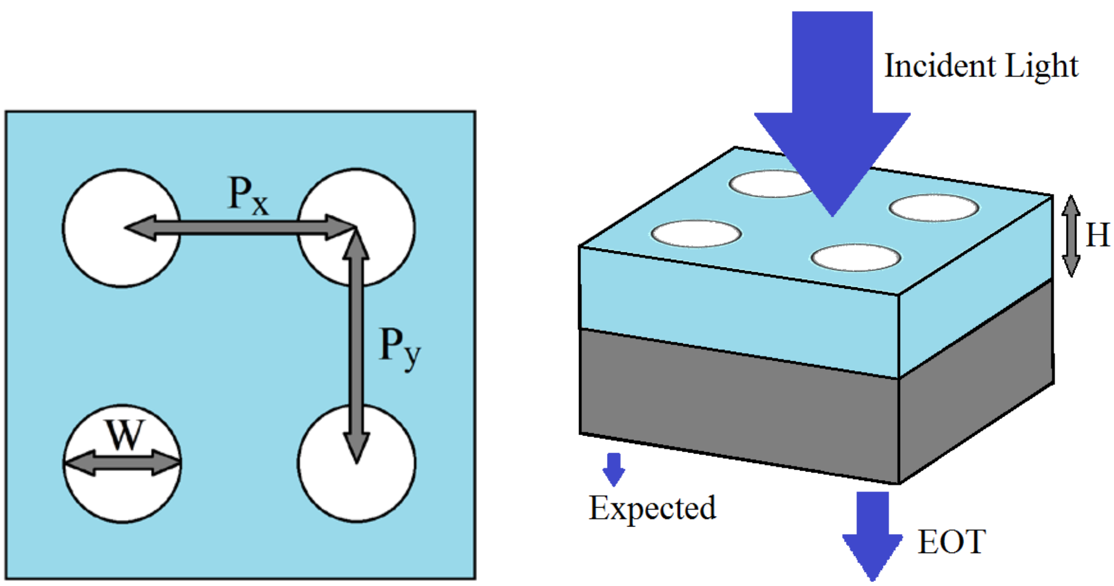

Figure 1: (left) Circular hole array structure in the ZnGaO film and (right) the overall structure with arrows indicating incident light and possible transmission cases.

2. ANALYTICAL AND NUMERICAL CALCULATIONS

Surface plasmon polaritons are bound electromagnetic waves that propagate along the interface between a dielectric and a conductor. Wavelength-dependent complex optical constants of conducting regions are utilized to analytically and numerically investigate the SPP excitation conditions with the 2D-hole array structures. Prior work has characterized a nominal $Zn_{0.974}Ga_{0.026}O$ film via IR-ellipsometry and Hall measurements with Drude parameters being extracted [**22**]. The doping concentrations of these films are ~10^{21} cm^{-3}.The same Drude formulation and extracted parameters was used here to determine the frequency-dependent optical constants for the ZnGaO film.

In order to determine the excitation condition for the investigated structures analytically, we first must find the real part of the SPP index. This is found according to [**28**]

$$n_{SPP} = \mathrm{Re}\sqrt{\frac{\varepsilon_d\, \varepsilon_c(\lambda)}{\varepsilon_d + \varepsilon_c(\lambda)}}\,, \tag{1}$$

where ε_d and ε_c are the electric permittivities of the dielectric and conductor respectively, with the latter taken to be complex. The coupling condition between the *x*-polarized incident light and a SPP-mode is satisfied when [**6**]

$$n_{SPP} = \lambda\sqrt{\left(\frac{n_d}{\lambda}\sin\theta + \frac{m}{P}\right)^2 + \left(\frac{n}{P}\right)^2}\,, \tag{2}$$

where the incident is at an angle θ from surface normal, P is the grating period assumed here to be uniform in each direction, n_d is the index of the dielectric region where the SPP mode is excited, m and n are all positive and negative integers (0, ±1, ±2...), λ is the incident wavelength which is equal to the excitation wavelength for a specific mode. In most cases where the dielectric is air and for normal incidence, Eq. 2 reduces to

$$n_{SPP} = \frac{\lambda}{P}\sqrt{m^2 + n^2}\,. \tag{3}$$

The incident medium of air is the case for the measurements and used for simulations condcuted in this work.

Figure 2 presents the analytic SPP index dispersion (dashed curve) according to Eq. 1 and several possible modes, as indicated with colored lines the right-most line showing the lowest order mode. The modes are calculated according to Eq. 3 for periods of 6 and 10 μm. In Fig. 2 the dispersion illustrates the typical inflection at the plasma wavelength, which for this material is λ ~1.4 μm as reported in Ref. [**22**]. The modes plotted illustrates up to m = n = 2 for non-degenerate cases (m,n = ±1,0 and 0, ±1 all give the same solution according to Eq. 3) . The crossing of the dispersion and a particular mode curve indicates the wavelength of corresponding excitation. The lowest mode order in each of these cases occurs practically at wavelengths equal to, albeit slightly larger than the period of the structure. As the period is increased, the excitation wavelength is increased accordingly.

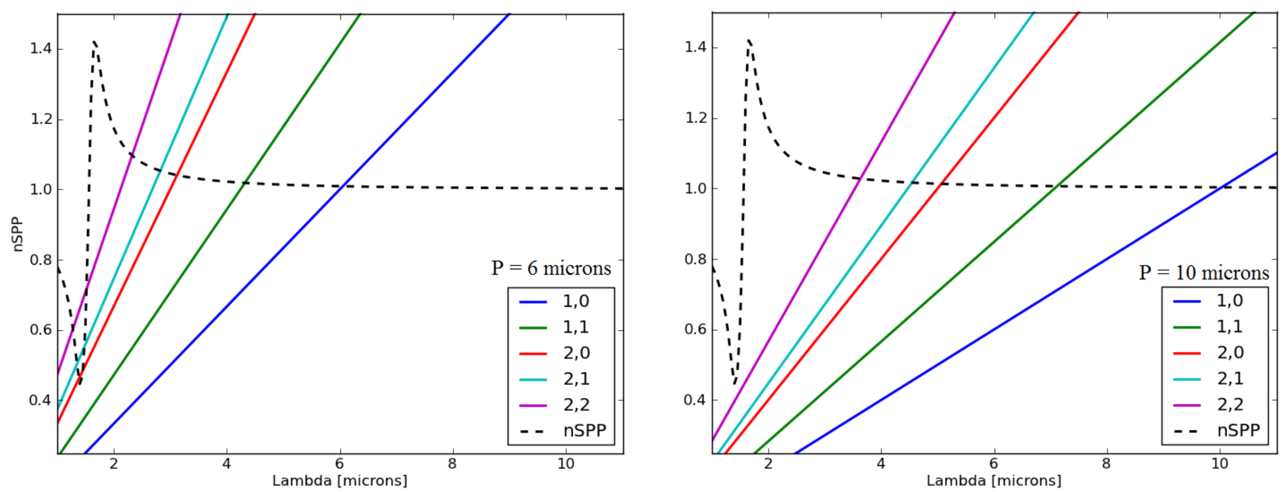

Figure 2: Analytic SPP (Eq. 1) and mode (Eq. 3) dispersion curves on ZnGaO structures with 6 μm (left) and 10 (right) μm periods. The modes (lines) plotted from right to left along the dashed line in each figure are 1,0, 1,1, 2,0, 2,1, and 2,2.

Lumerical FDTD, numerical finite difference time domain software, was used to model the electromagnetic transmission and reflection from the structure in Fig. 1. Simulations are completed for a structure with a simple

nonabsorbing Si substrate and a 1 μm thick conductive layer on top. This layer was defined as a material with the same Drude formulation discussed earlier for ZnGaO. The 2D-array was designed as periodic circular holes with a period and diameter of 6 and 3 μm respectively. The results of such simulations are presented in Figure 3 for incident light, linearly polarized in the *x*-direction. The plasma wavelength of the film, as previously noted, is ~1.4 μm, indicating that in the range of simulations, the film is expected to be reflective. The 1,0 and 1,1 modes are clearly apparent as both a dip in reflection and peak in transmission which is recognized as EOT. The wavelengths of these mode orders agree with those calculated analytically for SPP's in Fig. 2. This simulation provides proof of principal of EOT on doped ZnO 2-D structures as well as a means to optimize such structures in the future.

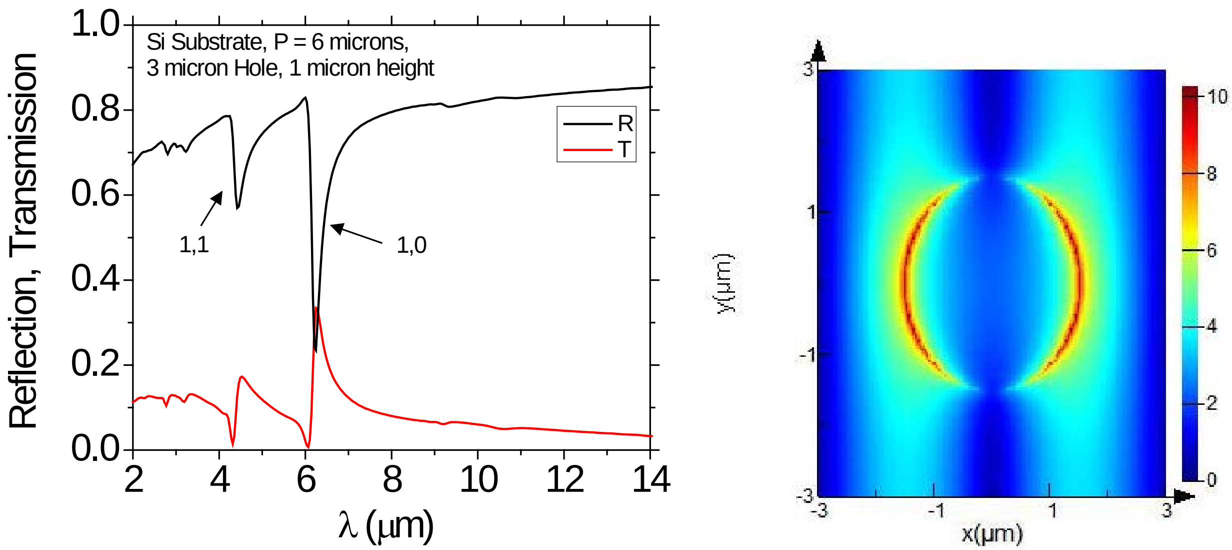

Figure 3: (left) Reflection and transmission as calculated numerically for a 2D circular hole array in highly conductive ZnGaO. Two mode orders are indicated. (right) Spatial electric field of the 1,0 mode order just above the film surface at the wavelength of reflection minima, λ = 6.25 μm . The film is 1μm thick, and the holes have periodicity and diameter of 6 and 3 μm respectively.

3. FABRICATED STRUCTURES

ZnO films, doped with Ga, were deposited on sapphire substrates via pulsed laser deposition (PLD) using a KrF excimer laser. The film, nominally $Zn_{0.974}Ga_{0.026}O$, was deposited in pure argon from a ZnO target with 3 wt.% Ga_2O_3. This method has been investigated recently for fabrication of highly conductive ZnO [**25, 29**]. The nominal film composition is determined from donor concentrations assuming all donors were contributed from Ga doping. [**30**]. The film in this work is ~ 500 nm thick, and the deposition parameters are similar to those reported and characterized in Ref. [**22**].

The film was then patterned into a 2D array of circular holes with periods of 6, 8, and 10 μm with varying hole apertures. The holes were ICP-etched for 10 minutes at 450 Watts using a BCl_3-Ar gas mixture. Figure 4 presents a scanning electron microscope (SEM) image of the structure and an energy dispersive X-ray (EDX) spectrum collected in one of the holes of the structure. The SEM image for this 10 μm period sample indicates that circular holes have been etched with some residual ZnO forming a ring around the surface. The EDX data, measured at the bottom of an etched hole, identifies aluminum while also having a relatively large zinc peak. This indicates that the electron beam is penetrating to the sapphire substrate but the ZnGaO film hasn't been completely etched through. The structures measured here then are circular conductive ZnO hole-arrays on a thin film of the same material. The etching process is still being optimized.

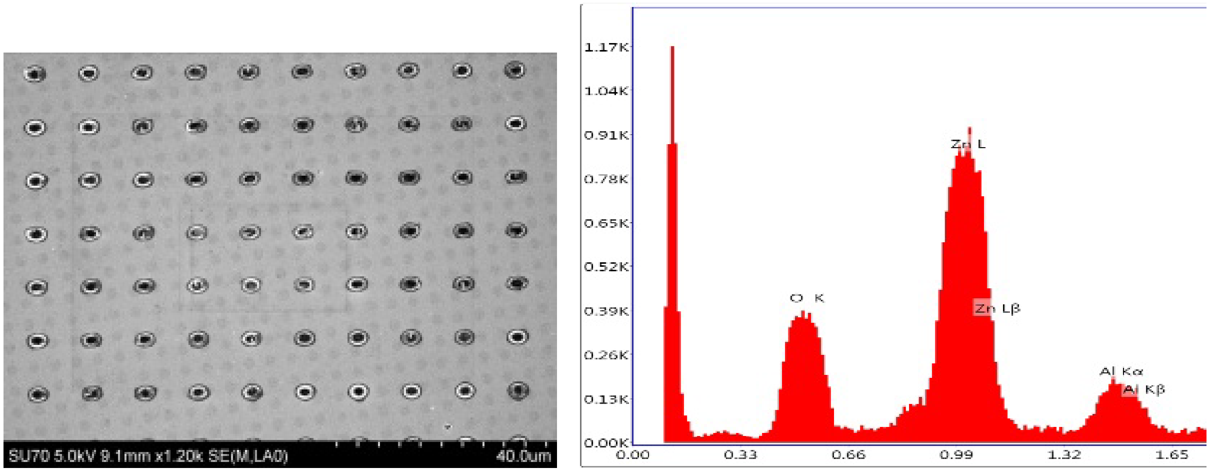

Figure 4: (left) SEM image of a patterned 2D circular hole array with 10 μm period. (right) EDX spectrum taken in one the etched holes indicating that some of the ZnO film remains between the holes and sapphire substrate.

4. RESULTS AND ANALYSIS

Figure 5 (left) presents the *x*-polarized reflection spectra measured using a Bruker Vertex 80v FTIR coupled with a Hyperion microscope for circular holes of 2 μm diameter. The corresponding spectra of the 2D arrays with periods of 6, 8, and 10 μm are shown with black, red, and blue curves respectively. The frequencies of the strongest and likely lowest order mode for periods of 6, 8, and 10 μm are 9.2, 10 and 10.4 μm respectively with weaker higher order modes also observed. Figure 5 (right) plots the experimentally observed resonance wavelengths of the 2 and 3 μm diameter hole structures and strongest mode (circles) and weaker higher order mode (triangles). The resonance wavelengths for the structures with the two different hole diameters qualitatively match. The noted absorption features, observed as dips in the reflection data, redshift with increasing period as expected from theory. These experimental data shows a noticeable redshift in resonance wavelength when compared to earlier analytical theory and numerical calculations (Figs. 2-3). It is noted that the angle of incidence however is non-normal and is actually a focused light-cone that is unavoidable in the current microscope setup. The light cone is specified to range 12-24º from the surface normal.

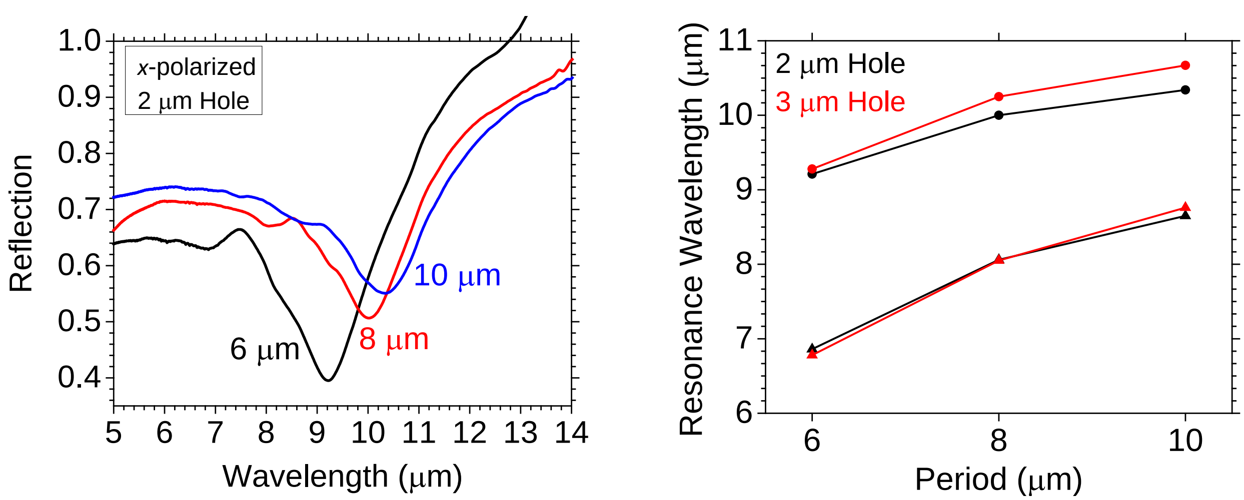

Figure 5: (left) Reflection spectra of 2D circular holes in ZnO film for three different pattern periods with the circular opening of 2 μm. (right) Resonance wavelengths experimentally determined from 2D hole array structures with 2 and 3 μm hole openings for periods of 6, 8, and 10 μm. The symbols represent (circles) the observed strongest feature and likely lowest order mode, and (triangles) a weaker, higher order mode.

In order to further investigate the effects of the angle of incidence, we calculate dispersion curves using Eqs. 1-2, the results of which are presented in Figure 6. All parameters are the same as those used in Fig. 2 except now the angle of incidence is 19º. It is noted that this still remains a simplification of the experimental angles expected. Traveling around the light-cone with reference to the sample plane, one can expect the experimental angles (keeping in mind the polarization still being in the *x*-direction) to range from positive 24º to negative 24º from normal incidence. The inclusion of this angle in the *x*-direction allows previously degenerate m^{th} orders to become non-degenerate. For example, the *m,n* mode negative 1,0 now is separate from the positive 1,0 resonance mode. To keep the plot simple, only mode orders of -1,0, and 1 are included. The -1,0 and 1,0 mode are pushed to longer and shorter wavelengths respectively than that of normal incidence (Fig. 2). The longest wavelength mode shown here is ~7.8 μm which is still shorter than the experimentally measured 9.2 μm (Fig. 5) for the 6 μm period hole structure. Profiles at some points also indicate there may be photoresist left on the surface of the structure which could serve as a partial high index incident dielectric region. Testing with Eq. 2, indicates this has the effect of further red-shifting modes, as would happen also if the excitation occurred on the substrate side of the structure. The microscope light cone and possible resist left on the surface are likely contributing factors to red-shifting of the experimental modes observed. While the experimental results demonstrated in this work utilize sapphire substrates we are undergoing a transition to highly resistive silicon substrates, which will enable EOT measurements in the LWIR where sapphire is no longer transparent.

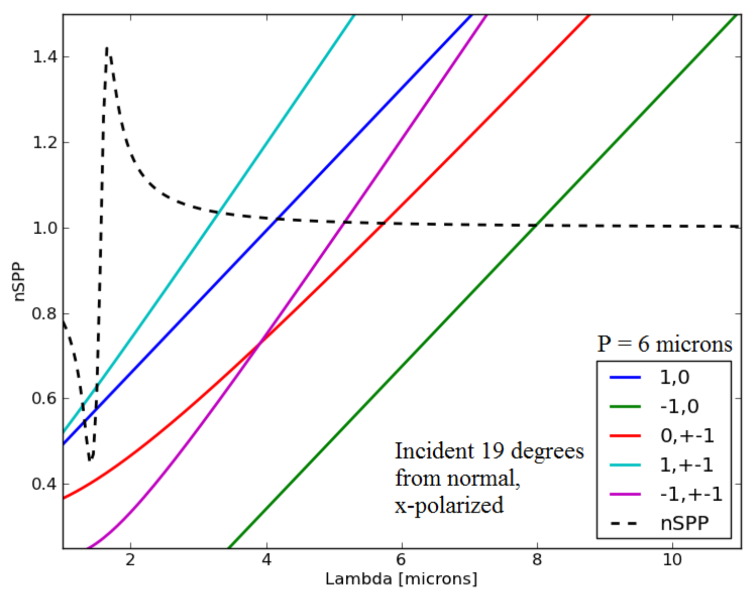

Figure 6: Analytic SPP (Eq. 1) and mode (Eq. 2) dispersion curves on a ZnGaO structure with a period of 6 μm, the polarization in the *x*-direction and a 19º angle of incidence. The "+-" in the legend indicates mode orders that are degenerate. The modes (lines) plotted from right to left along the dashed line in the figure are -1,0, 0,±1, -1,±1, 1,0, and 1,±1.

5. SUMMARY AND OUTLOOK

Circular hole array structures have been fabricated in highly conductive ZnGaO films on sapphire substrates. Reflection measurements on hole structures show strong resonances that increase in wavelength with increasing period. These experimental results are in qualitative agreement with analytical calculations, indicating the excitation of LWIR SPPs on the investigated ZnGaO 2D-hole-array structures. The development of suitable etch processing is ongoing with future films to be fabricated on highly resistive silicon substrates. Numerical results shown here for such structures indicate that EOT is possible and in agreement with known analytical theories.

ACKNOWLEDGMENTS

JWC and NNE would like to acknowledge support from the Air Force Office of Scientific Research (Program Manager Dr. Gernot Pomrenke) under contract number 12RY10COR while JRH would like to acknowledge support under LRIR No. 12RY05COR. DCL would like acknowledge support also from the Air Force Office of Scientific Research (Program Manager Dr. James Hwang) under grant number FA9550-10-1-0079.

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